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Absolute cross section for electron-impact ionization of He($1s2s\ ^3S$)

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We present an experimental determination of the electron-impact ionization cross section of the $1s2s\ ^3S$ state of helium, for which there is a serious long-lasting discrepancy between theory and experiment. A technique for the production of a fast, intense beam of helium in the $1s2s\ ^3S$ state only has been developed for this purpose, based on photodetachment of the He^- anion. The cross section is measured using the animated crossed beam technique. The present results are much lower than the experimental data of Dixon *et al.* [J. Phys. B **9**, 2617 (1976)] and are in excellent agreement with the calculation of Fursa and Bray [J. Phys. B **36**, 1663 (2003)].

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Helium is considered a benchmark for the study of electron correlation and, as such, has been the subject of much investigation. Although processes involving the ground state are now well understood, this is not always the case for the first excited state, He($1s2s\ ^3S$). For the particular case of electron-impact ionization, there has been only one experiment spanning a significant energy range, performed more than 40 years ago [1] and whose results are in sharp disagreement with the most accurate calculations performed over the past 20 years [2–5]. The difference of up to 60% in the total cross section is not acceptable as metastable helium plays an important role in a wide range of environments, from plasma physics to Bose-Einstein condensates [6], and there has consequently been many calls for further experimental investigation [2,3,5,7–9]. This discrepancy also raises uncertainties on theoretical values of the cross sections for higher-lying excited states, which are in great demand since no experimental cross sections are available.

He($1s2s\ ^3S$) lies 19.8 eV above the ground level and is the longest-lived neutral excited atomic state, with a lifetime of 7870 s [10]. Alpha particles are a natural product of controlled thermonuclear fusion and metastable helium is expected to play an important role in fusion devices due to its large excitation energy and ionization cross section. Pure helium plasma is also used for testing purpose and injection of neutral helium is a tool for temperature and electron density diagnostics based on detailed radiative-collisional models which require accurate atomic data [11–14].

Transfer of the He($1s2s\ ^3S$) large internal energy through binary collisions leads to Penning ionization of other atoms, molecules, or solids in a very efficient way. This process is therefore expected to be important in the chemistry of atmospheres and interstellar medium [15]. It also responsible for sustaining cold atmospheric pressure helium plasmas and creating molecular radicals in the plasma jet, a technique now applied for, e.g., skin treatment and disinfection [16–18]. The high reactivity of metastable helium confers it very low penetration depth and thus makes it an ideal candidate for surface treatment and analysis, which has spurred the fields of helium nanolithography [19,20] and metastable deexcitation microscopy [21,22]. Finally, metastable helium atoms can be easily cooled, manipulated, and detected, and have thus given rise to a variety of cold atoms experiments [6], including Bose-Einstein condensates [23], velocity selective coherent population trapping [24], and atomic Bragg scattering [25].

Measuring the electron-impact ionization of metastable helium is challenging, which explains the absence of other measurements since the work of Dixon *et al.* [1]. Difficulties that must be addressed are twofold: first, the source of metastable helium must have a high purity while keeping sufficient brightness; second, the measurement of absolute cross section is very sensitive to experimental inaccuracies. Over the past few decades, we have developed the animated crossed beam technique and the corresponding experimental setup, extensively described elsewhere [26,27], that allows accurate and absolute values of cross sections for electron-impact ionization of atoms and molecules to be measured. A source of pure metastable helium has been specifically designed for the experiment, as no existing source of fast metastable atoms matches the present requirements. Cross sections for electron-impact ionization of He($1s2s\ ^3S$) have been measured for electron kinetic energies ranging from threshold (4.77 eV) to 1000 eV.

The production of helium atoms in the $1s2s\ ^3S$ state is challenging since optical dipole excitation from the ground state is forbidden. Electron-impact excitation of ground-state helium is widely used, either in a gas discharge or using an electron gun [28,29], which produces a beam of ground-state helium containing minute fractions of singlet ($1s2s\ ^1S$) and triplet ($1s2s\ ^3S$) metastable atoms. Neutralization of fast He^+ ions onto alkali-metal vapor produces fast atoms in the $1s^2\ ^1S$, $1s2s\ ^{1,3}S$, and $1s2p\ ^{1,3}P$ excited states with a population distribution that has been characterized for all alkali-metal targets [28,30–34]. While generally acceptable, such a distribution is inappropriate when turning to state-specific measurements. We propose instead to use photodetachment of $He^-(1s2s2p\ ^4P^o)$, which leaves helium in the $1s2s\ ^3S$ state, provided that the photon energy is tuned below the $1s2p\ ^3P$ threshold, i.e., below 1.1 eV (see, e.g., Xi and Froese Fischer [35]).

The practical implementation of the state-specific production scheme follows three steps, represented in the sketch of the experimental setup shown in Fig. 1. First, He^+ ions are extracted from a duoplasmatron source fed with He gas and accelerated to 8 keV. Second, the He^+ beam is passed through a Na vapor cell where it is converted into He^- by double charge exchange with sodium atoms with an efficiency of the order of 1%. Downstream, a magnet selects the He^- component of the beam and injects it into the electron-impact ionization

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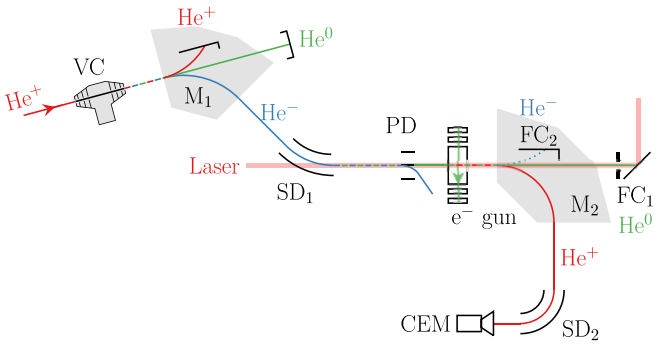

FIG. 1. Experimental setup. VC: sodium vapor cell; M_1: magnet mass selector; $SD_{1,2}$: spherical deflector; PD: planar deflector; M_2: magnetic analyzer; $FC_{1,2}$: Faraday cup; CEM: channel electron multiplier. When the planar deflector (PD) is off, He^- ions fly to the Faraday cup FC_2, as indicated by the dotted line. Only the major parts of the apparatus are shown here; for a more detailed sketch of the electron-impact apparatus (starting after M_1), see, e.g., Ref. [27].

setup. Although $He^-(1s2s2p\,^4P)$ is metastable, the lifetimes of its $J = 1/2$, $3/2$, and $5/2$ fine-structure components are sufficiently long (7.8 μs, 12.3 μs, and 359.0 μs, respectively [36]) to permit the use of conventional beam transport and detection techniques. In a third step, after passing through several deflectors, the anion beam interacts collinearly with the light from a CO_2 laser. Photodetachment occurs along this path (17.5 cm) and leaves helium atoms in the $1s2s\,^3S$ state only.

The CO_2 laser generates 10 W of light with $\lambda = 10.6$ μm and an M^2 factor smaller than 1.2. It is loosely focused onto the atomic beam by the combination of a divergent ($f = -10$ cm) and a convergent ($f = 20$ cm) lens, at mid-distance between the exit of the spherical deflector SD_1 and the electron beam. The optics were chosen so that the spot size (1.3 mm) is close to the atomic beam diameter (2 mm) and the Rayleigh range (10.4 cm) is large enough to maintain sufficient intensity throughout the detachment region, resulting in large detachment efficiencies. The laser beam enters the vacuum chamber through a laser window, reaches the atomic beam *via* a hole drilled in the spherical deflector SD_1 and leaves the vacuum chamber through another laser window in order to be collected by a power meter.

Before entering the collision region, the atomic beam is cleaned of the remaining negative ions by a planar deflector (PD). It is then intersected perpendicularly by the ribbon-shaped electron beam of the cold-cathode electron gun. The collision region is brought to 1000 V so that the He^+ ions formed by electron-impact ionization have a kinetic energy of 9 keV compared to the 8 keV energy of He^+ ions produced by collision with the residual gas up- and downstream. The ions produced by electron impact are selected by the analyzing magnet M_2, subsequently deflected by a 90° spherical deflector (SD_2), and counted with a channel electron multiplier (CEM).

The remaining metastable atoms fly straight through the magnet and are collected onto a Faraday cup (FC_1). The Faraday cup consists in a polished aluminum surface, oriented at 45° with respect to the direction of the incident beams, and a guard electrode, set to a high positive voltage. The inclination of the surface and the 95% reflectivity of aluminum for $\lambda = 10.6$ μm allows the laser beam, which still overlaps the metastable atom beam, to be reflected outside of the vacuum chamber. The current of neutral atoms is determined by measuring the current of secondary electrons emitted upon atom impact on the Al surface and extracted by the guard electrode. The secondary electron emission coefficient γ, necessary for determining the neutral current, is calibrated by measuring the attenuation of the He^- beam for varying laser powers. We found $\gamma = 3.44$ in average, with very little variation (3%).

The metastable atom source described above performs well, with He^- currents of 15 nA being routinely achieved in the collision region, and yielding currents of metastable helium, as measured in amperes, of 6 nA. This corresponds to neutral particle densities of 5×10^3 cm^{-3}. The detachment efficiency of 40% is very high and can be explained by two factors. First, the photodetachment cross section of He^- is large, reaching 10^{-16} cm^2 at the CO_2 laser wavelength [35]. Second, the photodetachment rate is inversely proportional to the photon energy which, for $\lambda = 10.6$ μm (0.117 eV), is 20 times lower than for visible light (500 nm, 2.48 eV). Therefore, all other quantities being equal, the detachment efficiency is strongly enhanced when using long-wavelength infrared radiation. Although free from other excited states, the metastable beam is contaminated by ground-state atoms due to spontaneous detachment and stripping on residual gas. Spontaneous detachment occurs along the 17.5-cm-long interaction region, located 2 m after the Na vapor cell, and neutralizes 2.3%, 1.7%, and 0.08% of the incoming anions initially in the $J = 1/2$, $3/2$, and $5/2$, respectively. Neutralization of the incoming He^- thus occurs at the level of 1%, assuming that the fine-structure states of He^- are statistically populated during the charge exchange process. Stripping collisions with the residual gas convert another 3.5% of the incoming anion beam into ground-state helium.

The atom source now well characterized, we can turn to the electron-impact ionization cross-section measurement. The cross section is measured using the animated cross beam method of Defrance *et al.* [26], whose main advantage lies in the fact that the form factor, related to the interaction volume, does not need to be determined (see, e.g., [27,37] for more details). In a nutshell, the ribbon-shaped electron beam is swept vertically across the atom beam in a linear see-saw motion while the He^+ ions produced are counted and the electron beam current I_e and secondary electron current I_s are measured. The total number K of He^+ ions produced during one sweep of the electron beam is related to the cross section σ by

$$\sigma = \frac{v_e v_n}{\left(v_e^2 + v_n^2\right)^{1/2}} \frac{\gamma e^2}{I_s I_e} \frac{u K}{\eta}, \tag{1}$$

where v_e and v_n are the velocities of the electrons and atoms, respectively, e is the elementary charge, u is the sweeping speed, and η is the detection efficiency. The sweeping speed u of $\sim$8 m s^{-1} is determined by measuring the travel time of the electron beam between two wires located above and below the atomic beam, and separated by a known distance. The detection efficiency for 8 keV He^+ ions is 0.95 ± 0.05, as estimated from the known detection efficiency for protons at 5 keV [38].

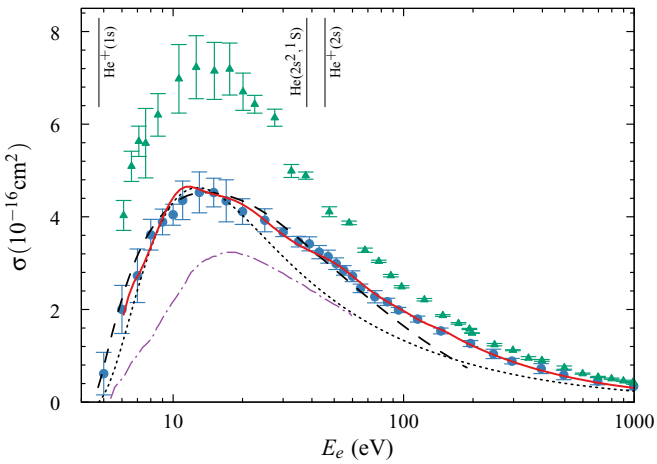

FIG. 2. Electron-impact ionization cross sections of He($1s2s\ ^3S$). Full circles: present work; full triangles: absolute measurement of Dixon *et al.* [1]; dotted line: CCC calculation [4]; dashed line: TDCC calculation [2]; dashed-dotted line: RMPS calculation [5]; full line: CCC and Born calculation [3]. The vertical lines indicate the position of the various thresholds. The error bars are the 2σ statistical uncertainties.

Two corrections to Eq. (1) must be considered in order to account for spurious experimental effects. First, some anions are detached *inside* the spherical deflector SD_1, where the neutrals produced do not have the correct trajectory to reach the collision region. By modeling the laser-atom overlap, we have determined that the measured γ must be multiplied by 1.03 in order to take this effect into account. Second, electron-impact ionization of ground-state atoms contaminating the metastable beam will also contribute to the He^+ signal. By performing a measurement when the laser was switched off, we could determine that the ground-state contribution amounts to less than 1% of the measured ionization yield. Uncertainties arising from systematic effects are estimated to be of the order of 8% and predominantly arise from the 5% uncertainty on the detection efficiency and the 6% uncertainty on the secondary electron emission coefficient.

The results for electron-impact ionization of He($1s2s\ ^3S$) are presented in Fig. 2 along with existing theories and experiment. Only one absolute measurement is available over a wide electron energy range, by Dixon *et al.* [1], and it lies much higher than the present results. In their experiment, a fast beam of metastable atoms was first formed by charge exchange between fast He^+ ions and cesium vapor and subsequently crossed perpendicularly by an electron beam. The cross section was determined under the assumption that, after charge exchange, 80% of the beam is in the $1s2s\ ^3S$ state, while the remaining atoms are in the ground state. This assumption was later weakened by Reynaud *et al.* [30] and Neynaber and Magnuson [31] who showed that, for a 1 keV He^+ beam, the neutral beam emerging after charge exchange consists in 60.5% of He($1s2s\ ^3S$), 23.5% of He($1s2s\ ^1S$) and 16% of He($1s^2\ ^1S$). We have therefore computed a rough estimate of the corresponding correction to the 4 keV data of Dixon *et al.* by using the 1 keV populations and the ionization cross section of He($1s2s\ ^1S$) from Ralchenko *et al.* [4]. The correction is found to be small, the larger ionization cross section of He($1s2s\ ^1S$) counterbalancing the reduced He($1s2s\ ^3S$) fraction, and certainly cannot account for the large discrepancy observed with the present experiment. The origin of such a large difference remains unclear.

A handful of absolute experiments were performed (see [39] and references therein) prior to the experiment of Dixon *et al.* [1]; however, they are limited to electron energies ranging from the metastable ionization threshold (4.77 eV) to the ground-state ionization threshold (24.59 eV). Indeed, the thermal-energy beams used contained predominantly ground-state atoms and therefore ionization of the small fraction of metastable atoms was masked by ground-state ionization above 24.59 eV. The cross sections obtained vary greatly in shape and magnitude (see, for example, Long and Geballe [39]) and vastly differ from the present cross section.

Before turning to comparison with theory, we may stress that, in both the present experiment and that of Dixon *et al.*, the measured signal is the sum of three contributions: (i) ionization of the outer electron leaving the He^+ ion in its ground state; (ii) ionization of the inner electron leaving the He^+ ion in an excited state; (iii) excitation to doubly excited states of He and subsequent autoionization.

On the theoretical side, the electron-impact ionization of He($1s2s\ ^3S$) is a benchmark process that has been the object of a number of theoretical calculations of ever-increasing sophistication. Numerous examples of model calculations can be found in the literature, such as Born-type and binary-encounter calculations, Deutsch-Märk formalism, or distorted wave calculations [1,2,7,40–44]. A series of *ab initio* calculations has also been performed in recent years, the results of which are represented in Fig. 2. Several convergent-close-coupling (CCC) calculations were dedicated to computing the electron-impact ionization cross section [3,4,8], and systematically fall more than a factor 1.5 below the experimental data of Dixon *et al.* [1]. In order to assess the validity of the CCC calculations, *R*-matrix method with pseudostate (RMPS) and time-dependent close-coupling (TDCC) calculations were also performed [2,5]. The present measurement lies higher than the RMPS calculation of Bartschart [5], but matches the TDCC calculation of Colgan and Pindzola [2] up to electron energies of 100 eV and is in excellent agreement with the calculation of Fursa and Bray [3] over the whole energy range covered. Fursa and Bray performed a frozen-core CCC calculation which was smoothly scaled, at higher energies, by the ratio between multicore and single-core Born calculations in order to account for ionization with excitation and excitation to doubly excited states of helium followed by autoionization, up to $n = 4$. While ionization through doubly excited states is negligible for ground-state atoms [45], the present measurement suggests along with [3] that it contributes to the ionization cross section for excited states. Surprisingly, the most recent frozen-core CCC calculation of Ralchenko *et al.* [4] deviates from the frozen-core CCC calculation of Fursa and Bray [3] at higher energies.

In summary, we have measured the cross section for the electron-impact ionization of He($1s2s\ ^3S$). A source capable of producing an intense, fast, and pure beam of He($1s2s\ ^3S$) atoms was designed and characterized, and can in principle be easily scaled up for applications requiring larger yields and densities. It may also pave the way to measurements of other quantities,

such as the photoionization cross section of He($1s2s\ ^3S$). The measured cross section for single ionization by electron impact is in excellent agreement with the calculation of Fursa and Bray [3], and lies much lower than previous experimental data [1]. The present results confirm the influence of doubly excited states in the process and suggest that such effects should be included in future accurate reference data [4].

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